# chemical dynamics versus transport dynamics in a simple model

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**Abstract**

Reaction equations of homogeneously mixed pollutants in the atmosphere can lead to non-stationary periodic solutions. It is important to know in which respect these solutions are modified under the influence of the atmospheric currents. We investigate this question in a very simple model: The reaction equations are modeled by the equations of the brusselator and the currents are represented by an isolated vortex. In the limit of high vortex currents we find again the homogeneous solutions whereas for smaller currents complicated spatial and temporal patterns emerge. The role of the diffusion as a singular perturbation is investigated.

## 1 INTRODUCTION

The concentrations of pollutants in the atmosphere depend on the atmospheric currents and the reaction equations between the pollutants. (Moreover they depend on cloud formation, humidity, ice etc, influences that will not be discussed here.)

When dealing with this problem the currents are usually replaced by their average and mixing is modeled by introducing 'turbulent diffusion'[1][2][3][4]. The problem of such approximations is that the chemical reactions depend on the *local* concentrations and not on averaged ones. A further disadvantage of this scheme is that the inserted turbulent diffusion is orders of magnitudes

higher than the molecular diffusion. This is questionable because diffusion is a singular[5][6] and sensitive perturbation as will be shown below.

If the atmospheric currents lead instantaneously to a homogeneous mixing of the pollutants, all concentrations are obtained from the chemical reaction equations alone. The solutions of these reaction equations need not approach a time-independent limit (fixed point for the dynamical system) but periodic fluctuations are possible as has recently been shown in a model system containing six pollutants and two pollutant sources[7][8][9]. A particular feature was the result that pollutant concentrations may change by an order of magnitude within a few days.

The solutions of the reaction equations become perturbed and then modified as soon as the atmospheric currents do not lead to a complete mixing any more. This situation could be modeled by adding to the reaction equations the above mentioned averaged currents plus a turbulent diffusion term that is sufficiently high. But according to the remarks made earlier a more appropriate approach is to assume that the diffusion is weak and to incorporate the transport equations in the reaction equations directly.

The temporal evolution of concentrations of $n$ reactants are described by the reaction-advection-diffusion equations:

$$\frac{\partial}{\partial t} c_i = f_i(c_1..c_n, \mathbf{x}, t) - \mathbf{v}(\mathbf{x}, t) \cdot \nabla c_i + \epsilon_i \Delta c_i, \quad i = 1, .., n, \qquad (1)$$

where the functions $f_i$ describe the chemical reactions and pollutant sources. These functions depend explicitly on the spatial coordinate $\mathbf{x}$ if the reactant $i$ is produced by a localized source. The second term represents the advection of pollutants by the velocity field $\mathbf{v}(\mathbf{x}, t)$ and the last one is the diffusion term. The coefficient $\epsilon$ is small and in a first approximation we neglect the diffusion. Later on we will investigate the addition of diffusion in our model very carefully.

In general the equations eq.(1) are rather involved. In this paper we choose the simplest possible system describing nontrivial reactions between constituents subjected to a simple current which is a solution of Euler's equation[10]. The reaction equations are taken from the brusselator[11]. Depending on the parameters the concentrations converge to a fixed point or limit cycle in the homogeneous case. Typically periodic solutions with sharp peaks occur. They are analogous to those obtained in the model of [7] where the chemistry of tracer constituents in the troposphere were described.

For maintaining the chemical reactions a 'pollutant source' is required. We choose a point source pouring out pollutants of sort $X$ placed in the velocity field of a two-dimensional isolated point vortex. The pollutant $X$ decays,

partly into harmless substance $E$ partly into a second constituent $Y$ that autocatalytically reacts with $X$ again according to the well known reactions of the brusselator

$$
\begin{aligned}
X &\to E \\
X &\to Y \\
2X + Y &\to 3X
\end{aligned}
\tag{2}
$$

Since molecular diffusion does not lead to a spreading on a macroscopic scale the vortex distributes the pollutants along the circular streamline containing the point source and thus the model becomes one dimensional. We note that this advection-reaction problem can be seen as an extension of the simple brusselator containing it as a limit limiting case for high vortex strengths (or when the source is close to the vortex center).

Next we discuss the distribution of concentrations on the circle. When the time $T$, needed for one circulation is small, the solutions become very similar to the reaction equations with a homogeneously distributed source. On the other hand, if $T$ is larger than a threshold value for an observer moving with the fluid, the distribution becomes periodic with period $T$, whereas for an observer at rest the distribution is stationary. When $T$ decreases the distributions become rather different: Time dependent solutions with period $nT$, $(n = 2, 3...)$, quasiperiodic and chaotic solutions are detected. All these lead to time dependent distributions that are infinitely degenerate on the circle and therefore depend on the initial distribution. Even if that is smooth, steps in the distributions occur, in the chaotic case on *each scale*. Therefore diffusion is a singular perturbation and switching it on along the streamline leads to drastic effects. For the period $2T$ case, the addition of diffusion can be understood completely. We find two time scales. The first, which is of the order of $1$[1] leads to coarse graining on the order of $\sqrt{\epsilon}$, the second which is of the order of $e^{\beta/\sqrt{\epsilon}}$, $\beta = \mathcal{O}(1)$ removes all the degeneracies of the solutions leading to a distribution that is smooth apart from one step. This step is intrinsic and would appear for a source of finite size along the streamline as well.

For parameters, that without diffusion lead to periodic solutions of higher period we observe pattern formation and spatiotemporal chaos. All these solutions have nothing in common with the case of homogeneous mixing we started with.

In section II we present the model, in section III we discuss its properties without diffusion, in section IV we concentrate on the role of diffusion. The conclusion ends the paper.

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[1] In this paper we use non-dimensional units.

## 2 THE MODEL

The reaction processes, eq.(2), lead to the well known reaction equations, which in non-dimensional form read as

$$\dot{c}_1 = c_1^2 c_2 - (1+b)c_1 \tag{3}$$
$$\dot{c}_2 = bc_1 - c_1^2 c_2$$

Here $c_1$ and $c_2$ are the concentrations of constituent $X$ and $Y$ respectively. The parameter $b$ presents the ratio between the decay rate of $X$ into harmless substances and the decay rate of $X$ into pollutant $Y$.

Without a source term the concentrations tend to zero. In our model we assume for simplicity a point like pollutant source that is fixed, but encounters the fluid field of a two-dimensional isolated vortex. This vortex produces a circular flow with velocity of modulus $\Gamma/r$ at distance $r$ from the center, $\Gamma$ being the strength of the vortex. This problem is one-dimensional since the chemical reactions can only take place along the circular streamline containing the source and can be mapped to the unit interval with periodic boundary conditions. Thus the flow is completely represented by one parameter, the dimensionless velocity $v$. We obtain the following combined reaction transport equations:

$$\frac{\partial}{\partial t}c_1 = s\delta(x) + c_1^2 c_2 - (1+b)c_1 - v\frac{\partial}{\partial x}c_1 + \epsilon\frac{\partial^2}{\partial x^2}c_1 \tag{4}$$
$$\frac{\partial}{\partial t}c_2 = bc_1 - c_1^2 c_2 - v\frac{\partial}{\partial x}c_2 + \epsilon\frac{\partial^2}{\partial x^2}c_2$$

where $s$ is the strength of the source located at $x = 0$. We allow here for a very small amount of diffusion with diffusion constant $\epsilon$.

Apart from the diffusion constant $\epsilon$ the equations contain three relevant parameters, the decay ratio $b$, the source strength $s$ and the velocity $v$.

Eq.(4) represent the Eulerian description of an observer at rest. If we change to the Lagrangian description (i.e. to an observer moving with the fluid) eq.(4) is transformed into

$$\frac{\partial}{\partial t}c_1 = s\sum_{n=0}^{\infty}\delta(\bar{x} + vt + n) + c_1^2 c_2 - (1+b)c_1 + \epsilon\frac{\partial^2}{\partial \bar{x}^2}c_1 \tag{5}$$
$$\frac{\partial}{\partial t}c_2 = bc_1 - c_1^2 c_2 + \epsilon\frac{\partial^2}{\partial \bar{x}^2}c_2$$
$$\bar{x} = x - vt \bmod 1$$

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The solutions to this model will be discussed in the following two sections.

## 3   PROPERTIES OF THE MODEL WITHOUT DIFFUSION

First let us discuss the transport reaction equations, eq.(4) and eq.(5) with diffusion switched off:

For $\epsilon = 0$ the coordinate $\bar{x}$ can be interpreted as a parameter equivalent to a time translation $\bar{x}/v$ in the driving term. Therefore in the moving frame we have to solve for any $\bar{x}$ the *ordinary* differential equations

$$\frac{d}{dt}c_1 = s \sum_{n=0}^{\infty} \delta(\bar{x} + vt + n) + c_1^2 c_2 - (1+b)c_1 \tag{6}$$

$$\frac{d}{dt}c_2 = bc_1 - c_1^2 c_2$$

The spatial distribution of the concentrations is completely determined by the solution of the above equation and the initial distribution. Obviously we have to take into consideration the time shift of the driving for different points.

In eq.(6) the difference to the 'usual' brusselator is the periodic $\delta$ function time dependence of the source. Thus we have a periodically driven (kicked) brusselator with one extra parameter, the period of the driving $T \equiv 1/v$. The periodically driven brusselator has been investigated in different contexts considering a constant plus a sinusoidal or delta function time dependence of the source [13][14][15]. In the $T \to 0$ limit the normal brusselator is recovered, i. e. very frequent injections correspond to an almost uniform source. In this limit the parameter plane $s - b$ can be divided into two regions (fig.1): For values corresponding to higher source strengths the concentrations converge to the fixed point $c_1^* = s, c_2^* = b/s$. As $s$ is decreased, the fixed point becomes unstable and a Hopf bifurcation occurs along the curve $s = \sqrt{b+1}$ forming the boundary between the two regions. Below this curve the system converges to a limit cycle, i. e. the concentrations oscillate periodically. (fig.2.)

As the periodic driving is switched on $0 < T \ll 1$, a periodic pulsation with period $T$ of the concentrations appears. Moreover the initially two dimensional phase space becomes three dimensional by including the cyclic variable $t/T \bmod 1$ due to the driving. Thus the dimensionality of the attractor increases as well, and the original fixed point turns to a limit cycle, representing a periodic time dependence, but still remains a fixed point of the stroboscopic map. Similarly, the original limit cycle becomes either a torus corresponding to a quasiperiodic dynamics with one of the periods equal to $T$ (fig.3) or a periodic orbit with large period. This is expected from the characteristic features of periodically driven oscillators. There exist resonant regions for driving

frequencies close to their natural frequency multiplied by a rational number. These resonant regions appear here below the Hopf bifurcation curve. In the parameter space of these regions Arnold tongues are detected analogous to those of the so called 'circle map'[16] (fig.4,5). As another effect of the driving the Hopf bifurcation curve moves to smaller values of $s$ as $T$ increases (fig.1). Since the dynamics is given by a set of two non-autonomous ordinary differential equations chaotic behavior is also possible for some values of the parameters leading to a strange attractor in the stroboscopic section (fig.6).

Next we consider the *distribution* of the concentrations due to the properties of the driven system. Let us discuss the typical cases:
$\alpha$)*periodic time dependence with period $T$*:
This behavior occurs for large $T$, i.e. as the velocity of the flow becomes small. In this case the concentrations oscillate and the phase of the oscillations is determined by the phase of the driving $t/T \, mod \, 1$. Thus the final state does not depend on the initial conditions. In the moving frame the only difference in the periodic time dependence at different points of the flow is a time lag $\bar{x}T$.

$$c(\bar{x}, t) = c(0, t + \bar{x}T). \tag{7}$$

If we move back to the standing frame ($\bar{x} \to x - vt$) we obtain asymptotically a stationary state $c(x, t) = c(0, xT)$ i.e. the concentrations converge to a $x$-dependent function constant in time.

$\beta$) *periodic oscillations with period $nT$*:
This behavior corresponds to the resonant regions. In this case the concentrations can take $n$ different values for a given phase of the driving depending on the initial conditions.

$$c(\bar{x}, t) = \{c(0, t + iT + \bar{x}T)\}, \quad i = 0...n - 1, \tag{8}$$

The boundary between the basins of attraction of the $n$ branches of the solution is a twisted (Mobius-like) surface so that the basin of attraction of branch $i$ becomes the basin of attraction of branch $i + 1$ mod n after one period $T$. Thus any smooth initial condition must have at least one intersection with this surface. At this point the concentrations converge to two different branches so a discontinuity appears in the spatial distribution of the concentrations (fig.7). Note that this step is not a consequence of the delta function in eq.(6) but is due to geometrical constraints. An initially random distribution can lead to a completely staggered distribution whose envelopes are the $n$ branches of the solution. In the moving frame the steps remain at the same position $\bar{x}$.

$\gamma$) *quasiperiodic time-dependence*:
This is present in a region below the Hopf bifurcation curve between the

resonances and is pronounced for small $T$ because with increasing $T$ the size of the region below the Hopf curve shrinks and at the same time the resonant islands grow in size. This case corresponds to a motion on a torus in the phase space. The dynamics can be characterized by two cyclic angle-like variables, one of them is the phase of the driving and the other one depends smoothly on the initial concentrations. $c(\bar{x}, t) = c(\bar{x}, t + \tau(\bar{x})) = c(t + \tau(\bar{x}) + \bar{x}T)$ Therefore an initially smooth distribution remains smooth in $\bar{x}$ for all times (except at the initial position of the source where the time lag of the driving by $T$ leads to a discontinuity.)

$\delta$) *chaotic time-dependence*:
In this case the time dependence is very sensitive to the initial conditions and thus the distribution becomes irregular on each scale regardless how smooth the initial distribution may have been (fig.9a).

## 4   THE ROLE OF DIFFUSION

Without diffusion the final distributions (except those with the period $T$) have infinite degeneracy due to an arbitrary uneven [2] number of steps. Therefore diffusion is a *singular* perturbation that has significant consequences for the system as small as $\epsilon$ may be. For the following computations we used the Crank-Nicholson scheme combined with operator splitting [12].

We discuss here the simplest nontrivial case of eq.(8) first, which occurs for period $2T$. We denote with a $-+$ $(+-)$ step an 'upward' ('downward') steep increase (decrease) of the concentration, but exclude the strong increase of $c_1$ at the location of the source. If the diffusion is small enough we can treat a step as isolated (for a very long time). Due to diffusion the step will move with a drift velocity, cf (fig.7). Scaling and symmetry arguments suggest that it should be proportional to a higher power of $\sqrt{\epsilon}$ and in fact we find numerically a dependence $\propto \epsilon$. The important point, however, is that, averaged over $2T$, each isolated step moves with the *same* drift velocity. In fact after time $T$ a $-+$ step becomes a $+-$ step and vice versa. What we expect then as the essential ingredient of eq.(8) is that $f$ tries to enforce solutions with period of $2T$. In appendix A we have derived a simple function $\mathbf{f}$ that has just this property *and* makes it possible to treat eq.(8) analytically. Then we find: first, for times

$$T_1 = \mathcal{O}(1) \tag{9}$$

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[2]   we do not count the strong increase of the $c_1$ concentration due to the $\delta$ function shape as a step.

all steps with distance of $\mathcal{O}(\sqrt{\epsilon})$ vanish. During this time the diffusion does nothing but a coarse graining. Second, over a period of about

$$T_2 = e^{\beta/\sqrt{\epsilon}}, \ \beta = \mathcal{O}(1) \tag{10}$$

all other steps are effected. The diffusion removes the degeneracies by and by, until a final state emerges that has no steps at all, besides the generic one that cannot be removed. This state has global stability in our model. Numerically we find the same phenomena for $\mathbf{f}$ of the brusselator, cf. fig.7b.

The effects described here occur quite independently from how small $\epsilon$ is again demonstrating that the diffusion is a singular perturbation. On the other hand $T_2$ depends exponentially on $1/\sqrt{\epsilon}$. When a further perturbation has to be added acting on a time scale $\tau$ we expect quite different situations depending on whether $T_2 > \tau$ or $T_2 < \tau$. This means that the effect of such a perturbation depends *sensitively* on $\sqrt{\epsilon}$ which shows that introducing diffusion as a parameter - as has been done by introducing 'turbulent diffusion' - is quite a dangerous approximation.

We expect even more complicated properties of the concentrations having higher periods (in absence of diffusion). There are two reasons for that: i) if the period is $nT$ the system has at any location $n-1$ choices for the height of a step, ii) the steps are no longer equivalent but are separated in classes and only steps within the *same* class change into each other and therefore move with the same mean drift velocity $v_d$. Indeed, the effect of diffusion on the periodic solution can be very significant in some cases by leading to a complicated irregular behavior of the system in space and time. As can be seen from fig.8, inside the chaotic concentration field coherent regions with regular periodic time dependence appear and disappear continuously. This kind of spatiotemporal intermittency has been observed in different extended systems[17])[18]) (e.g. in case of coupled maps[19])[20][21]. If one starts the simulation with a smooth initial distribution first at least the intrinsic step appears as described above. The pertubation of the periodic solution around the discontinuity leads to a chaotic time dependence which due to the diffusive coupling spreads over the whole system. Such behavior can be observed for parameters which lie in the vicinity of the chaotic regimes in the $\epsilon = 0$ case. The solution appears already for very small $\epsilon$ demonstrating again that diffusion is a singular perturbation.

In case of quasiperiodic local behavior, instead of a finite number of discrete branches, a continuous set of solutions exists filling the torus in the phase space. Thus the discontinuity present in the case without diffusion is easily removed by an arbitrarily weak diffusion leading to coherent quasiperiodic oscillations of the whole system.

When the parameters correspond to chaotic local dynamics, diffusion tends to

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form correlated regions of finite extent in space and time (fig.9) similar to the case above. As $\epsilon$ is increased, the local dynamics becomes completely regular with a frozen irregular distribution in space which certainly depends on the initial distribution.

## 5    Conclusion

High peaks can appear in periodic solutions of chemical reaction equations in which the constituents are *homogeneously* mixed tracer gases of the atmosphere. However, depending on the motion of the fluid the mixing need not be homogeneous at all, and the question arises how these solutions will then change.

In this paper we investigate this question for a simple model, the brusselator with pointlike source in a one-vortex flow. Simple as the model appears, it already demonstrates the strong modifications occurring as soon as we move away from the homogeneous situation. One observes this when computing the concentration distribution along the (closed) streamline in which the source is located. As a function of time we detect solutions that are very similar to those of the homogeneous case. This happens as long as the period $T$ of the flow is small. Furthermore we find solutions with period $nT$, moreover quasiperiodic and chaotic ones. All these solutions, except that with period $T$ are infinitely degenerate and therefore depend on the initial distribution. Even if that is smooth, the distributions can have asymptotically an arbitrary (uneven) number of discontinuities, in the chaotic case on *each scale*.

In such situations diffusion is a singular perturbation and switching on arbitrary small diffusion along the streamline has two effects: First after a time of $\mathcal{O}(1)$ it leads to a 'coarse graining' of the distribution on a space scale $\propto \sqrt{\epsilon}$ where $\epsilon$ is the strength of the diffusion. Second on a time scale $\propto e^{\sqrt{\alpha}/\sqrt{\epsilon}}$, $\alpha = \mathcal{O}(1)$ it removes all discontinuities but one for solutions which have (without diffusion) period $2T$. This shows that the solutions depend sensitively on $\sqrt{\epsilon}$. For parameters that lead (without diffusion) to solutions of higher period, quasiperiodic solutions and chaotic ones were observed, moreover pattern formation and spatio temporal chaos. All those solutions have nothing in common with the case of homogeneous mixing we started with.

Although this one-dimensional model is far from being a realistic representation of the chemistry and transport in the atmosphere, it shows that even a trivial non-turbulent flow interacting with a simple regular chemical dynamics of just two reactants can lead to a complex irregular behavior of the concentration fields.

## A    appendix

In this appendix we derive the properties of eq.(1) for our model assuming that without diffusion the solution has a $2T$ period in the moving system. We have

$$\partial_t \mathbf{c} = \mathbf{f}(\mathbf{c}, \bar{x}, t) + \epsilon \partial_{\bar{x}}^2 \mathbf{c} \tag{A.1}$$

with the periodic boundary conditions

$$\begin{aligned}
\mathbf{c}(\bar{x}, t) &= \mathbf{c}(\bar{x} + 1, t) \\
\partial_{\bar{x}} \mathbf{c}(\bar{x}, t) &= \partial_{\bar{x}} \mathbf{c}(\bar{x} + 1, t)
\end{aligned} \tag{A.2}$$

This equation holds true in the frame moving with a velocity $v = 1/T$.

The use of $\mathbf{c}$ can be awkward since the components have to be positive. Therefore we write

$$\mathbf{n} = \mathbf{c} + \mathbf{const} \tag{A.3}$$

and get the equation for $\mathbf{n}$

$$\begin{aligned}
\partial_t \mathbf{n} &= \mathbf{g}(\mathbf{n}, \bar{x}, t) + \epsilon \partial_{\bar{x}}^2 \mathbf{n} \\
&\quad with \\
\mathbf{g}(\mathbf{n}, \bar{x}, t) &= \mathbf{f}(\mathbf{const} + \mathbf{n}, \bar{x}, t)
\end{aligned} \tag{A.4}$$

and the periodic boundary conditions

$$\begin{aligned}
\mathbf{n}(\bar{x}, t) &= \mathbf{n}(\bar{x} + 1, t) \\
\partial_{\bar{x}} \mathbf{n}(\bar{x}, t) &= \partial_{\bar{x}} \mathbf{n}(\bar{x} + 1, t)
\end{aligned} \tag{A.5}$$

Without diffusion $\mathbf{n}$ moves exponentially fast to its asymptotic limit $\mathbf{n}^{(0)}$ having the properties

$$\begin{aligned}
\mathbf{n}^{(0)}(\bar{x}, t) &= \mathbf{n}^{(0)}(\bar{x}, t + 2T) \\
&\quad \text{and either} \\
\mathbf{n}^{(0)}(\bar{x}, t) &= \mathbf{n}^{(0)}(t - \bar{x}T) \\
&\quad or \\
\mathbf{n}^{(0)}(\bar{x}, t) &= \mathbf{n}^{(0)}(T + t - \bar{x}T)
\end{aligned} \tag{A.6}$$

$n^{(0)}$ and its properties remain important also if diffusion is switched on since $g$ can be expanded around $n^{(0)}$.

To understand the physics of eq.(A.4) with conditions eq.(A.6) we construct a simple model for the function $g$ in three steps:

<u>step 1</u>: we introduce a very simple $\mathbf{n}^{(0)}$

$$n_1^{(0)}(\bar{x}, t) = \Re\{ae^{i\pi(t/T - \bar{x})}\} \tag{A.7}$$
$$n_2^{(0)}(\bar{x}, t) = \Im\{ae^{i\pi(t/T - \bar{x})}\}$$

and we represent the two dimensional vectors by complex numbers.

<u>step 2</u>: we use the ansatz

$$n(\bar{x}, t) = e^{i\pi(t/T - \bar{x})} \cdot m(\bar{x}, t) \tag{A.8}$$

and get the pde for $m$

$$
\begin{aligned}
\partial_t m &= \tilde{g} - i\pi\epsilon\partial_{\bar{x}}m - \pi^2\epsilon m + \epsilon\partial_{\bar{x}}^2 m \\
&\quad with \\
\tilde{g} &= e^{-i\pi(t/T - \bar{x})}g(e^{i\pi(t/T - \bar{x})}m, \bar{x}, t) - (i\pi/T)m
\end{aligned}
\tag{A.9}
$$

The terms $i\pi\epsilon\partial_{\bar{x}}m$ and $\pi^2\epsilon m$ are of higher order in $\sqrt{\epsilon}$ and will be left out for simplicity. The boundary conditions of eq.(A.5) are replaced by

$$m(\bar{x}, t) = -m(\bar{x} + 1, t) \tag{A.10}$$
$$\partial_{\bar{x}}m(\bar{x}, t) = -\partial_{\bar{x}}m(\bar{x} + 1, t)$$

<u>step 3</u>: we construct a simple $\tilde{g}$. Because of eq.(A.7) and eq.(A.8) $m^{(0)}$ can take two values only,

$$m^{(0)} = \pm a \tag{A.11}$$

and $a$ can be chosen to be real and positive. When $m$ is in the neighborhood of $m^{(0)}$ $\tilde{g}$ can be expanded and we obtain

$$
\begin{aligned}
\tilde{g} &= -\alpha(\bar{x}, t)(m - a) + ... \\
&\quad or \\
\tilde{g} &= -\alpha(\bar{x}, t)(m + a) + ...
\end{aligned}
\tag{A.12}
$$

For $\alpha(\bar{x}, t)$ we insert a real positive constant[3]. The linear approximation of $\tilde{g}$ is of course incorrect if $m$ is not close to $\pm a$. A nonlinearity is added simply by the prescription

$$\tilde{g} = \begin{cases} -\alpha(m - a) \text{ for } \mid m - a \mid < \mid m + a \mid \\ -\alpha(m + a) \text{ else} \end{cases}$$

Thus we get the partial differential equation:

$$\begin{aligned} \partial_t m &= \tilde{g} + \epsilon \partial_{\bar{x}}^2 m \\ &\quad with \end{aligned} \tag{A.13}$$

$$\tilde{g} = \begin{cases} -\alpha(m - a) \text{ for } \Re m > 0 \\ -\alpha(m + a) \text{ for } \Re m \leq 0 \end{cases}$$

Boundary conditions are given by eq.(A.10). The connection between n and $m$ is given by eq.(A.8) and real and imaginary part of $n$ are the components of **n**.

**properties of the solutions of** eq.(A.13)

I) Diffusion switched off, i.e. $\epsilon = 0$,

$m$ consists asymptotically of an uneven number of steps with values $\pm a$. The number of steps can be arbitrarily high and is determined exclusively by the initial distribution of $m$.

II) Diffusion switched on, i.e. $\epsilon > 0$,

1) Isolated step

we assume that there is a constant velocity $\sqrt{\epsilon}w$ with which the step is moving. Transforming to new coordinates $y$ with

$$\bar{x} = y + \sqrt{\epsilon}wt \tag{A.14}$$

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[3]  $\alpha$ could be a complex constant as well as long as the real part is positive.

and assuming that the step occurs at $y = 0$ the two equations are to be solved:

$$0 = -\alpha(m + a) + \sqrt{\epsilon}w\partial_y m + \epsilon\partial_y^2 m, \ y < 0$$
$$0 = -\alpha(m - a) + \sqrt{\epsilon}w\partial_y m + \epsilon\partial_y^2 m, \ y > 0$$

because of the boundary conditions for the isolated step

$$m(-\infty) = -a$$
$$m(\infty) = a$$

the solution is

$$m_-(y) = Ae^{\gamma y/\sqrt{\epsilon}} - a, \ \gamma = (1/2)(-w + \sqrt{4\alpha + w^2}), \ y \leq 0 \qquad (\text{A.15})$$
$$m_+(y) = Be^{\tilde{\gamma}y/\sqrt{\epsilon}} + a, \ \tilde{\gamma} = (1/2)(-w - \sqrt{4\alpha + w^2}), \ y \geq 0$$

with the boundary condition

$$m_-(0) = m_+(0)$$
$$m'_-(0) = m'_+(0)$$

Because of eq.(A.13) there is the further condition

$$\Re\{m_-(0)\} = 0$$

Therefore $A = a$ and $B = -a$, $m$ is real and the condition for $w$ is obtained from

$$a\gamma = -a\tilde{\gamma}$$

which means

$$w = 0$$

2) two interacting steps isolated from the rest

Let the $-+$ step be left, the $+-$ step be right. Both steps move because of interacting with each other and we assume that the interaction changes speed and shape of the steps only slowly (the distance $2x_d$ between them decreases of course).

First we rescale to avoid the $\epsilon$ dependence

$$\xi = \frac{\bar{x}}{\sqrt{\epsilon}} \qquad (\text{A.16})$$

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Next we transform into a coordinate system moving with the $-+$ step, whose position is at $0$. we obtain

$$\xi = \eta + w_d t \tag{A.17}$$

and

$$0 = -\alpha(m_- + a) + w_d \partial_\eta m_- + \partial_\eta^2 m_-, \ \eta \le 0 \tag{A.18}$$
$$0 = -\alpha(m_+ - a) + w_d \partial_\eta m_+ + \partial_\eta^2 m_+, \ \eta \ge 0$$

Boundary conditions:

$$m_-(-\infty) = -a$$
$$m_-(0) = m_+(0) \tag{A.19}$$
$$m'_-(0) = m'_+(0)$$

Furthermore the presence of the $+-$ step is taken care of by the condition

$$m'_+(\xi_d) = 0 \tag{A.20}$$

and we have the constraint (cf eq.(A.13))

$$\Re\{m_-(0)\} = 0 \tag{A.21}$$

Then we get with an exponential ansatz (cf eq.(A.15)

$$m_-(\eta) = Ae^{\gamma\eta} - a, \ \eta \le 0, \tag{A.22}$$
$$m_+(\eta) = Be^{\tilde{\gamma}\eta} + Ce^{\gamma\eta} + a, \ 0 \le \eta \le \xi_d$$

Again $m$ can be chosen to be real and the conditions eq.(A.19), eq.(A.20) and eq.(A.21) yield

$$A - a = B + C + a$$
$$A\gamma = B\tilde{\gamma} + C\gamma \tag{A.23}$$
$$0 = B\tilde{\gamma}e^{\tilde{\gamma}\xi_d} + C\gamma e^{\gamma\xi_d}$$
$$A - a = 0$$

From these equations we get $w_d$ (neglecting all terms $w_d^2$ and higher)

$$w_d \approx 2\sqrt{\alpha}e^{-2\sqrt{\alpha}\xi_d} \tag{A.24}$$

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which is correct for

$$\sqrt{\alpha}\xi_d > 1 \qquad (A.25)$$

From scaling arguments we infer that eq.(A.25) gives the correct order of magnitude for $\sqrt{\alpha}\xi_d < 1$.

One can use eq.(A.24) also to prove that there is no stationary state. If it were all the equations were exact and in particular eq.(A.24) which in turn would be a contradiction.

Now we compute the lifetime $t_l$ of a step which is in the original coordinates

$$t_l = \frac{1}{4\alpha}(e^{2\sqrt{\alpha}x_d/\sqrt{\epsilon}} - 1) \qquad (A.26)$$

3) $n$ interacting steps

To treat this problem we take into account the interaction between nearest neighbors only ( the interaction between next nearest neighbors is exponentially small compared to the interaction between the nearest neighbors). Then it is sufficient to look into the problem of one step between two other steps. We approximate the interaction again by boundary conditions and obtain two conditions of the form eq.(A.20). Doing an analogous computation with the same approximations we obtain for the velocity of the step

$$w_d \approx 2\sqrt{\alpha}(e^{-2\sqrt{\alpha}x_{d+}/\sqrt{\epsilon}} - e^{-2\sqrt{\alpha}x_{d-}/\sqrt{\epsilon}}) \qquad (A.27)$$

Here $2x_{d+}$ $(2x_{d-})$ is the distance to the right (left) step. From this result we conclude that all states with more than one step will be unstable since two neighboring steps will annihilate each other.

**References**

[1] G.T. Csanady, turbulent diffusion in the environment, D. Reidel Publishing Company, Dordrecht 1973

[2] A.S. Monin and A.M. Yaglom, statistical fluid dynamics: mechanics of turbulence, volume 1, second edition, THE MIT PRESS, Cambridge, Massachusetts 1973

[3] S. Panchev, dynamic meteorology, D. Reidel Publishing Company, Dordrecht 1985

[4] K. Nester, H.-J. Panitz, F. Fiedler, Meteor. and Atmos. Phys. **57**, 201 (1995) and references therein

[5] A.M. Turing, Philos. Trans. R. Soc. London **B237**, 37 (1952)

[6] R. E. Bellmann, perturbation techniques in mathematics, physics and engineering, Holt, Rinehart and Winston, New York 1964

[7] D. Poppe and H. Lustfeld, J.Geophys. Res. **101**, 14373 (1996)

[8] M. Krol and D. Poppe, to be published in J.Atmos.Chem.

[9] H. Lustfeld, preprint

[10] Á. Péntek, T. Tél and Z. Toroczkai, J.Phys.A **28** (1995)

[11] H. Haken, synergetics, an introduction, Springer (1983)

[12] W.H. Press et. al., numerical recipies in C, Cambridge Univ. Press, (1992)

[13] I. Schreiber et.al., Phys. Lett. A **128**,66 (1988)

[14] T. Kai and K. Tomita, Prog. Theor.Phys. **61**, 54 (1979)

[15] D.G. Aronson et.al., Phys. Rev **A 33**, 2190 (1986)

[16] E. Ott, Chaos in Dynamical Systems, Cambridge Univ. Press, (1993)

[17] K. Kaneko, Prog. Theor. Phys. **72**, 480 (1984)

[18] H. Chatè, P. Maneville, Physica D **32**, 409 (1988)

[19] S.H. Kim and V. Hlavacek, Chem. Engng. Sci. **41**, 2767 (1986)

[20] I. Schreiber and M. Marek, Phys.Lett **A 91**, 263 (1982)

[21] G.V. Osipov, A.S. Pikovsky, M.G. Rosenblum, J.Kurths, Phys. Rev. **E 55**, 2353 (1997)

Fig. 1.  Curves corresponding to the Hopf bifurcation in the parameter plane $s - b$ for different values of the period $T$.

Fig. 2.  Constant in time and periodic behavior of the concentrations $c_1$ and $c_2$ for the unforced brusselator ($T = 0$). The parameters are $s = 2.5$, $b = 3.0$ and $s = 1.0$, $b = 3.0$, respectively.

Fig. 3.  Quasiperiodic time dependence of the concentrations $c_1$ and $c_2$ (a), and stroboscopic section (b) for $s = 1.0$, $b = 3.0$ and $T = 1.0$.

Fig. 4.  Stroboscopic plot of $c_2$ in function of $T$ for $s = 1.9$ and $b = 7.7$. The Hopf bifurcation occurs around $T = 1.03$ and there are resonant windows inside the quasiperiodic region labelled by the ratio of the two periods.

Fig. 5.  Periodic(blank) and quasiperiodic(gray) regions in a section of the parameter space for $T = 1.0$. The behavior of the system was identified by calculating the leading Lyapunov exponent which is smaller then $-0.0025$ for the blank region.

Fig. 6.  Chaotic time dependence of the concentrations $c_1$ and $c_2$ and the stroboscopic section of the strange attractor. The parameters are $s = 1.2$, $b = 7.0$ and $T = 1.36$.

Fig. 7.  Spatiotemporal plot of the concentrations $c_1$ (left) and $c_2$ (right) along the streamlines in the co-moving frame represented on a grayscale, so that concentrations increase from black to white. The simulation was started with both concentrations equal to zero and the initial position of the source is at $x = 0.2$. Parameters are $s = 1.0$ $b = 5.0$ and $T = 1.7$ In case (a) $\epsilon = 0$ and a non-moving discontinuity is present at $x = 0.2$. When diffusion is switched on $\epsilon = 0.001$ the discontinuity becomes rounded and moves (to the left in this case) along the streamline.

Fig. 8.  Stroboscopic spatiotemporal plot of concentration $c_2$ for parameters $s = 0.8$, $b = 6.0$ and $T = 1.85$, that correspond to a periodic behavior with period $3T$ when diffusion is neglected. Here $\epsilon = 2 \cdot 10^{-5}$ that leads to an irregular spatiotemporal dynamics.

Fig. 9.  Stroboscopic spatiotemporal plots of concentration $c_2$ for $s = 0.8$, $b = 6.0$ and $T = 1.89$. This parameters correspond to a chaotic local dynamics when diffusion is not considered. We assumed that the initial concentrations are randomly distributed in a small interval $[0, 0.0001]$ for both constituents. The diffusion coefficient is $\epsilon = 0$ (a), $\epsilon = 1.5 \cdot 10^{-5}$ (b) and $\epsilon = 2 \cdot 10^{-5}$ (c), respectively.
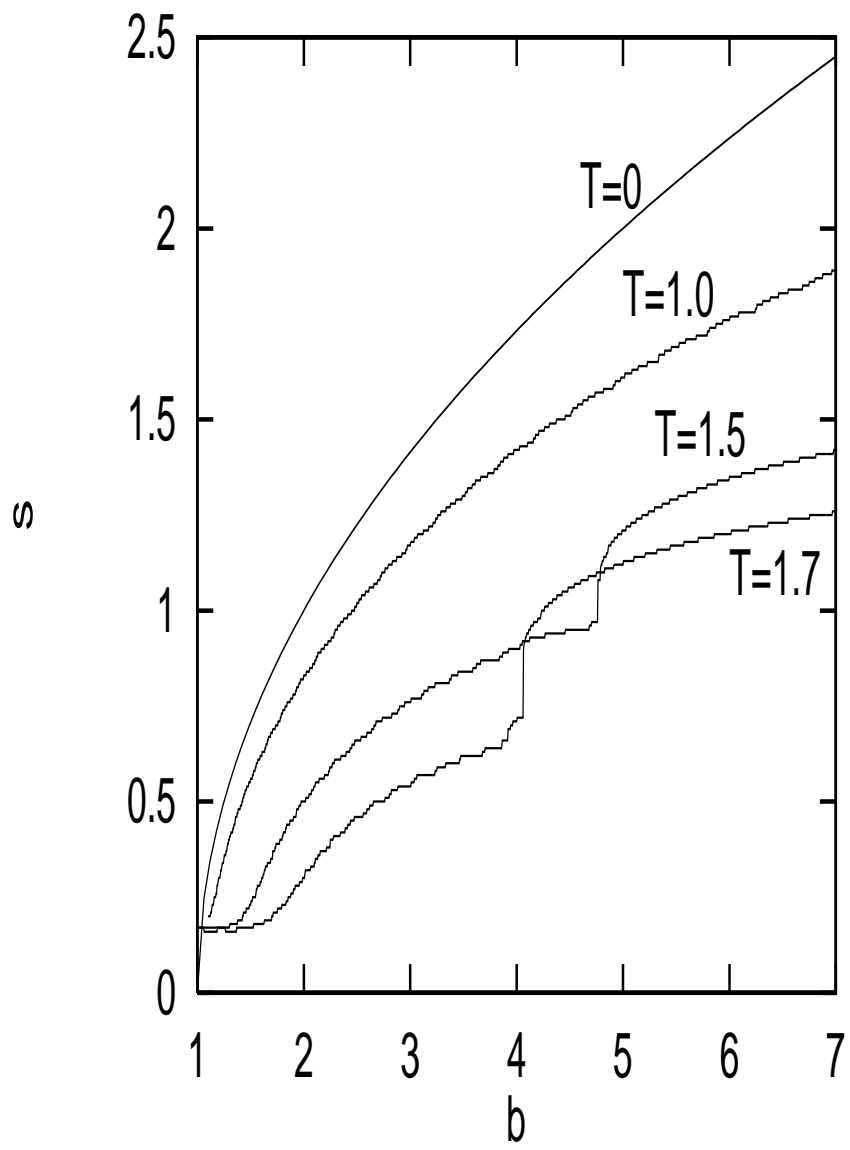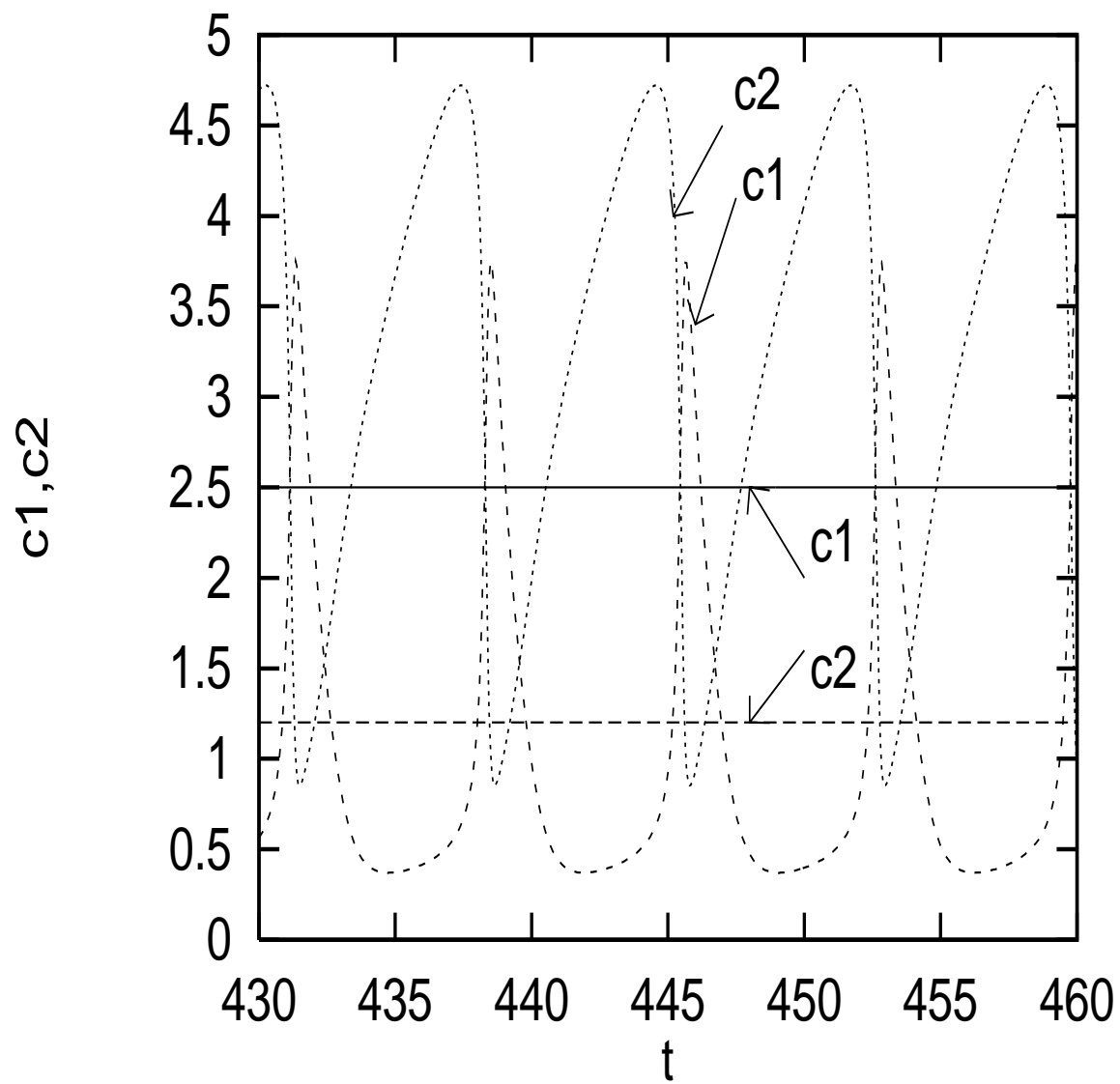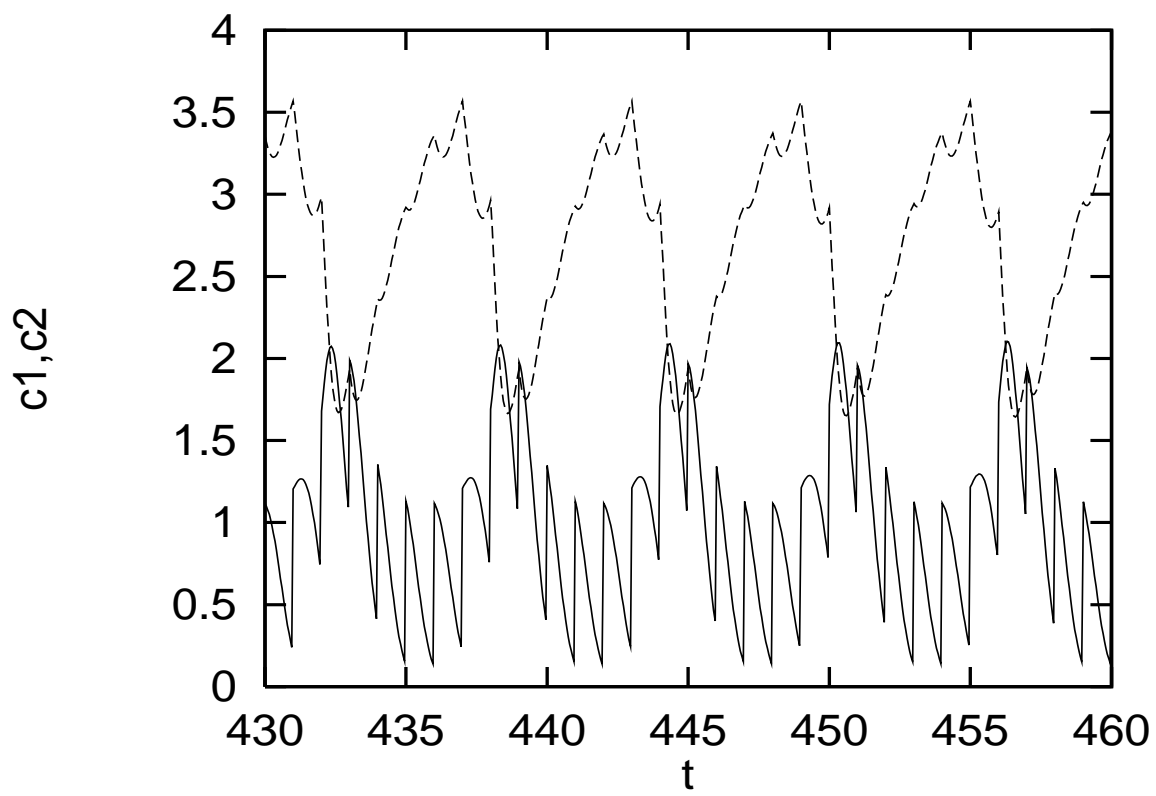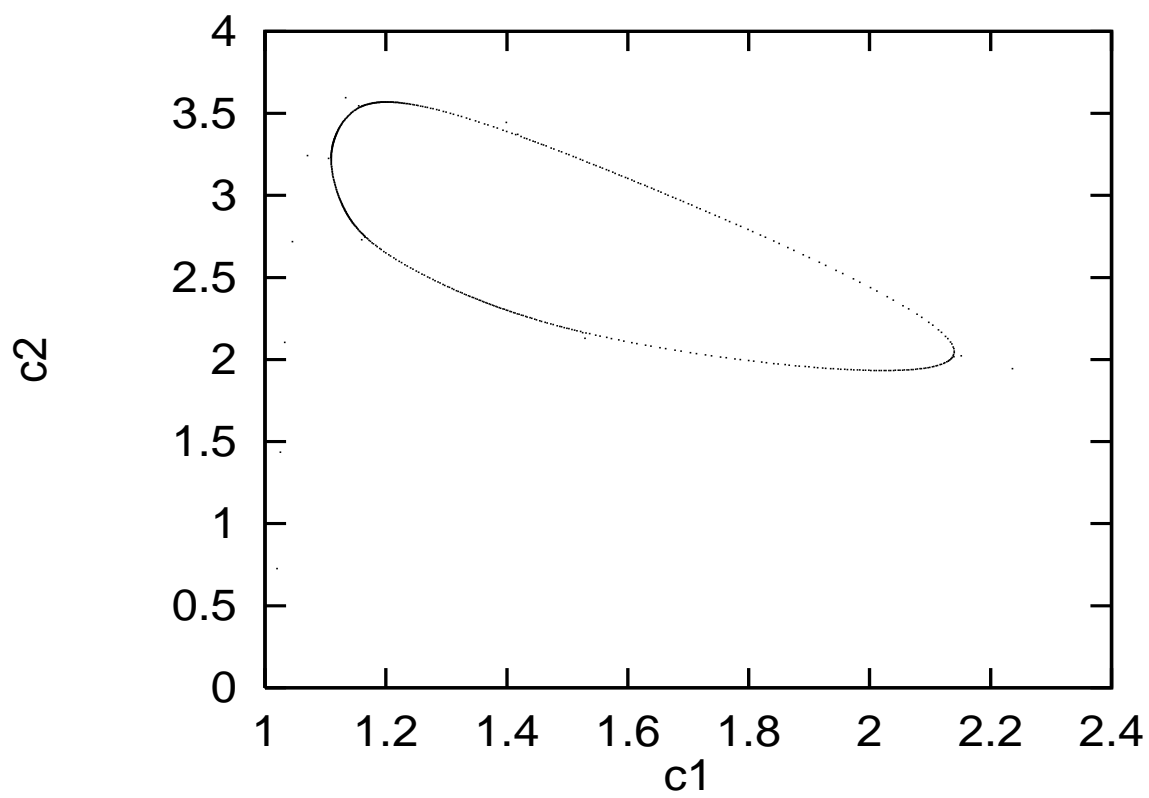
fig.1

19

fig.2

a)



b)

21

fig.4

fig.5

a)



b)

a)

b)

$\bar{x}$

0        c2        1

t

fig.8

26

a)



b)



c)

27